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Specification

1. Title of the Invention

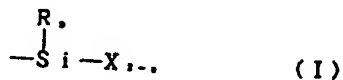
A curing composition and its application

2. Scope of Claims

(1) A curing composition in which the chief components are

(a) a hot-melt resin, and

(b) an oligomer of number average molecular weight in the range 500 to 50,000, having at least one hydrolyzable silyl group represented by general formula (I)



(where R represents a hydrogen atom or an alkyl group with 1 to 4 carbons, X represents a hydroxyl group or a hydrolyzable group and n is 0, 1 or 2) per molecule in a side chain and/or as a terminal of the main chain.

(2) A curing composition according to Claim (I) where, in general formula (I), the hydrolyzable group represented by X is a hydrogen atom, alkoxy group, acyloxy group, ketoxymato group, amino group, amido group, aminooxy group, mercapto group or alkenyloxy

group, and when there are two or more of X then these may be the same or different.

(3) A curing composition according to Claim (1) or (2) where the compounding ratio of the a) and b) components is a weight ratio in the range 100 : 5 to 10 : 100.

(4) A curing composition according to Claim (3) where component a) is a hot-melt butyl.

(5) A curing composition according to Claim (4) where the oligomer b) component is an isobutylene polymer.

(6) A hot-melt adhesive which employs a curing composition according to Claims (1) to (5).

(7) A vibration-deadening/damping material which employs a curing composition according to Claims (4) or (5).

3. Detailed Description of the Invention

[Industrial Field of Application]

The present invention relates to a curing composition and its application; more specifically, it relates to a curing composition of improved heat resistance and markedly improved vibration-deadening/damping characteristics, and to a hot-melt adhesive or a vibration-deadening/damping material employing this curing composition.

[Prior Art]

Hot-melt resins are polymers which are solids or waxes at room temperature. When heated so that their temperature lies in the range about 100-250°C, they flow and, in the case where used as an adhesive, they wet and adhere to surfaces of various kinds.

However, since a hot-melt resin comprises a system which is employed based on the introduction and removal of heat, when it is used for example as an adhesive, there are limits to the heat resistance following adhesion. Consequently, in fields where there is a strong demand for heat resistance, while a hot-melt resin may look promising in terms of its usability there are limits to the employment thereof, and raising the heat resistance has been regarded as a problem of considerable importance.

Against this background, polyamide and polyester hot-melt resins have been marked to meet such a demand but, on account of their melt viscosity, the application temperature has to be raised and the problem arises that use is impossible with adherends which are susceptible to heat.

Polyester/polyether block copolymer (DuPont; Dyvax), polyester/polyamide block copolymer (Monsanto; Montac) and polyether/polyamide block copolymer (Atochem; Pebax) have been introduced as hot-melt polymers of improved heat resistance, but their melt viscosity is high and again in terms of cost they cannot be said to be practical.

On the other hand, methods have also been proposed, and have appeared on the market, for enhancing the heat

resistance by bringing about a crosslinking reaction by some means following melt application (an example being the 'reactive hot melts').

However, the means adopted in such methods cannot be said to be simple in that, in order to bring about crosslinking of the hot-melt resin itself, the hot-melt resin is subjected forcibly to modification and crosslinking sites are manifested by thermal or chemical means. For such reasons, the development of a curing composition where the heat resistance can be simply enhanced is awaited by the industry.

Furthermore, with regard to vibration-deadening/damping materials, the development of a material which satisfies the following requirements is also awaited.

A vibration-deadening/damping material is desired which has the softness of a rubber material and high energy-dissipation ($\tan \delta = 0.4$ or above) and where, in terms of overall temperature dependency, frequency dependency and displacement dependency, little change is exhibited in these characteristics.

However, the fact is that, as yet, no material has been obtained which is satisfactory from the point of view of properties, cost and usability.

For example, taking the case of an ordinary silicone rubber, while the temperature dependency thereof is low, $\tan \delta$ is also low at 0.05-0.1, so it cannot be employed as a vibration-deadening/damping material. Furthermore, taking the case of butyl rubber, $\tan \delta$ is high at 0.4 or above, so it appears a desirable material but there is

the problem that $\tan \delta$ falls considerably in the high temperature region. Thus, the development of a curing composition with improved vibration-deadening/damping properties is awaited by the industry.

Consequently, an objective of the present invention lies in providing a curing composition where a hot-melt resin may be used as it is without modification, where the heat resistance is simply raised and, furthermore, where the vibration-deadening/damping properties are markedly improved.

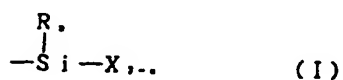
A further objective of the present invention lies in providing a hot-melt adhesive or a vibration-deadening/damping rubber material employing this curing composition.

[Means for Resolving the Problem]

The present invention has been made following painstaking research to resolve the aforesaid problems.

Specifically, the present invention is a curing composition in which the chief components are

- (a) a hot-melt resin, and
- (b) an oligomer of number average molecular weight in the range 500 to 50,000, having at least one hydrolyzable silyl group represented by general formula (I)



(where R represents a hydrogen atom or an alkyl group with 1 to 4 carbons, X represents a hydroxyl group or a

hydrolyzable group and n is 0, 1 or 2) per molecule in a side chain and/or as a terminal of the main chain.

The hot-melt resin which is the a) component used in the present invention is not particularly restricted and, normally, a commercial such hot-melt resin can be used like an EVA, polyamide, polyester, polyurethane, acrylic, butyl or polyolefin type hot-melt resin.

From the point of usability, it is preferred that its softening point be about 100-200°C but there is no restriction to this.

The hot-melt butyl used in the present invention is not particularly restricted and, normally, it is possible to use a commercial such material. This may be either material marketed as the resin alone or material marketed as a hot-melt butyl in which fillers, etc, have been compounded. Examples include butyl rubber of degree of unsaturation about 0.5-5.0 (IIR) and the chlorobutyl or bromobutyl Vistanex series (made by Exxon).

It is a feature of the present invention that the hot-melt resin or hot-melt butyl used may be employed as it is, without having been subjected to crosslinking and curing by vulcanization or the like.

The b) component employed in the present invention is an oligomer having at least one hydrolyzable silyl group represented by general formula (I) per molecule in a side chain and/or as a terminal of the main chain. The main chain from which the oligomer is composed is not particularly restricted and examples include a main

chain based on polyether, polyester, acrylic, vinyl ether, polycarbonate, polyisoprene, polyisobutylene or polybutadiene. However, in terms of the post-cure rubber elasticity not being impaired, it is preferred that it be non-crystalline at room temperature. From the point of view of usability and post-cure rubber elasticity, it is also preferred that the number average molecular weight of this oligomer lies in the range 500 to 50,000, and more preferably in the range 1,000 to 15,000.

In general formula (I), R represents a hydrogen atom or an alkyl group with 1 to 4 carbons. The alkyl group may be linear or branched, and preferred examples are the methyl group and ethyl group.

X represents a hydroxyl group or a hydrolyzable group. These are bonded to a silicon atom to form the hydrolyzable silyl group represented by (I).

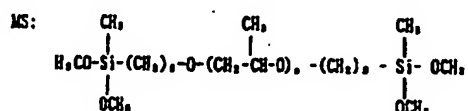
That is to say, reference to a hydrolyzable silyl group in this specification means a group comprising hydroxy or hydrolyzable groups bonded to a silicon atom, which can be hydrolysed by moisture in the presence or absence of a silanol condensation catalyst. Specific examples of the hydrolyzable groups are hydrogen atom, alkoxy group, acyloxy group, ketoximate group, amino group, amido group, aminooxy group, mercapto group, alkenyloxy group and other such generally-used groups. Alkoxy groups are particularly preferred from amongst these examples, in that they are easy to handle and hydrolysis conditions are mild. From one to three hydroxyl and/or hydrolyzable groups can be bonded to one silicon atom

and, where there are two or more thereof, these may be the same or different.

There may be one or more silicon atoms forming the hydrolyzable silyl group and in the case where silicon atoms are linked by means of siloxane bonds, or the like, up to 20 thereof are preferred.

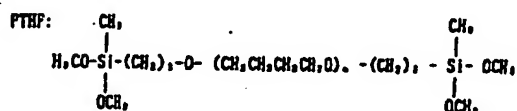
The following examples may be given of this kind of b) component.

Polyether type (polyoxypropylene)



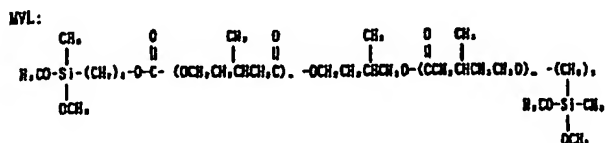
$$M_n \approx 8,000 \quad P_n(\text{Si}) \approx 1.4 \sim 1.8$$

Polyether type (polyoxybutylene)



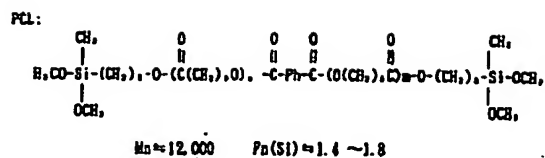
$$M_n \approx 4,000 \quad P_n(\text{Si}) \approx 1.4 \sim 1.8$$

Polyester type (polymethyl valerolactone)

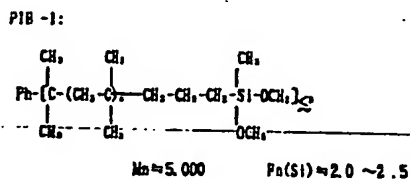


$$M_n \approx 4,000 \quad P_n(\text{Si}) \approx 1.3 \sim 1.7$$

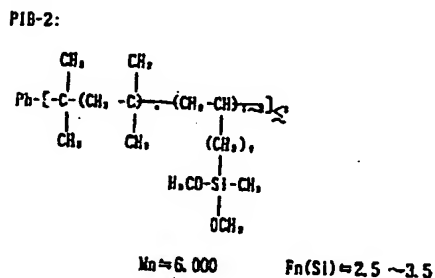
Polyester type (polycaprolactone)



Polyolefin type (polyisobutylene)



Polyolefin type (polyisobutylene)



In the combinations of the a) and b) components employed in the present invention, the materials described above can be used as appropriate but, from the point of view of a stable manifestation of the post-cure properties, it is preferred that there be employed a combination which exhibits comparatively good compatibility prior to curing. However there is no restriction thereto. This is because it is also possible to improve the compatibility sufficiently by using a plasticizer, or the like, along with the a) and b) components. Again, in the case where a hot-melt butyl is used as component a), both from the point of view of compatibility and also from the point of view of not impairing the properties of the hot-melt butyl itself, it is

particularly preferred that an isobutylene polymer with a polyisobutylene main chain be used as the b) component.

The compounding ratio of the a) and b) components is a weight ratio in the range 100 : 5 to 10 : 100. If there is less than 100 : 5 of component b), then there is little heat-resistance improvement or other effects due to the b) component. If the a) component content is less than 10 : 100, then the effect of the a) component in reducing the temperature dependency of $\tan \delta$, etc, is lowered.

In terms of specific applications, where the composition is used for a hot-melt adhesive it is preferred that the compounding ratio of the a) and b) components be a weight ratio in the range 100 : 5 to 10 : 100, and more preferably 100 : 10 to 50 : 100. If there is less b) component than this, then there is little heat-resistance improvement effect, while if there is too much, no further enhancement in heat-resistance improvement effect is seen and only the cost is raised, so this is undesirable.

Where used for a vibration-deadening/damping material, it is preferred that the compounding ratio of the a) and b) components be a weight ratio in the range 100 : 5 to 10 : 100, and more preferably 100 : 10 to 50 : 100. If there is less b) component than this, then shape retention at high temperature is difficult (there is a lowering of the elastic modulus), while if there is too much there is an increased lowering of $\tan \delta$ at high temperatures, so this is undesirable.

In the curing composition of the present invention, besides the a) and b) components, which are the chief components, there can of course be used, where required, various types of silane compound as agents for regulating properties. Furthermore, there can be optionally added various types of fillers, plasticizers, tackifying resins, the silanol condensation catalysts normally used for curing the oligomer with hydrolyzable silyl groups which constitutes the b) component, water, hydrates of inorganic compounds, ageing inhibitors, ultraviolet light absorbers, lubricants, pigments, foaming agents, adhesion conferring agents and the like.

Examples of the fillers which can be used in the present invention are wood flour, pulp, cotton⁴, asbestos, glass fibre, carbon fibre, mica, walnut shell flour, rice hull flour, graphite, diatomaceous earth, white clay, fumed silica, precipitated silica, silicic anhydride, carbon black, calcium carbonate, clay, talc, titanium dioxide, magnesium carbonate, quartz, microfine aluminium powder, flint powder, zinc powder or the like. These fillers may be used on their own or two or more may be used together.

Examples of the plasticizers are hydrocarbon compounds such as polybutene, hydrogenated polybutene, α -methylstyrene oligomer, biphenyl, triphenyl, triaryldimethane, alkylene triphenyl, liquid polybutadiene, hydrogenated liquid polybutadiene, alkyl diphenyl and partially hydrogenated terphenyl; chloro-paraffins; phthalate esters such as dibutyl phthalate, diheptyl phthalate, di(2-ethylhexyl)phthalate, butyl benzyl phthalate and butyl phthalylbutyl glycolate; non-aromatic dibasic acid esters such as dioctyl adipate and

dioctyl sebacate; polyalkylene glycol esters such as diethylene glycol benzoate and triethylene glycol dibenzoate; and phosphate esters such as tricresyl phosphate and tributyl phosphate. These may be used on their own or two or more can be used together.

In order to cure the oligomer component with hydrolyzable silyl groups, which is the b) component of the curing composition of the present invention, there can optionally be used a silanol condensation catalyst. Examples of such condensation catalysts are titanate esters such as tetrabutyl titanate and tetrapropyl titanate; carboxylic acid tin salts such as dibutyltin dilaurate, dibutyltin maleate, dibutyltin diacetate, tin octylate and tin naphthenate; the reaction products of dibutyltin oxide and phthalate esters; dibutyltin diacetylacetonate; organo-aluminium compounds such as aluminium tris-acetylacetonate, aluminium tris-ethylacetoacetate and diisopropoxyaluminium ethylacetoacetate; chelate compounds such as zirconium tetraacetylacetonate and titanium tetraacetylacetonate; lead octylate; amine compounds such as butylamine, laurylamine, monoethanolamine, triethylenetetramine, guanidine, 2-ethyl-4-methylimidazole and 1,3-diazabicyclo(5,4,6)undecene-7 (DBU), or the carboxylic acid salts of these; and also other known silanol catalysts such as acidic catalysts and basic catalysts, etc.

In the curing composition of the present invention there may be jointly employed various types of adhesion conferring agents for the purpose of further raising the adhesion properties. Specifically, by employing one or more from amongst epoxy resins, phenolic resins,

aminosilane compounds, epoxy silane compounds and other types of silane coupling agents, alkyl titanates and aromatic polyisocyanates, it is possible to enhance the adhesion properties in terms of various types of adherends.

[Examples]

Below, the curing composition of the present invention is explained in further detail by means of examples but the invention is not to be restricted in any way by these examples.

Examples 1 to 14

Curing compositions were prepared by mixing the a) and components b) together in the proportions shown in Table 1 and then, after filling moulding frames of thickness 3 mm at 80°C, hardening and curing were carried out for 1 week at 50°C. Next, after leaving for 1 hour at 150°C in each case, the softening state of the curing composition was observed by eye. These results are also shown in Table 1. In the results, where the shape was retained and there was no tack, this was indicated by the symbol O; where the shape was retained but there was slight tack, this was indicated by Δ; and where there was melting, this was indicated by X.

Now, in every case, 1% of each of the three types of ageing inhibitors Tinuvin 327, Sanol 770 and Irganox 1010, that is to say a total of 3%, was compounded in terms of component b).

Comparative Examples 1 to 6

The softened state at 150°C of the hot-melt resins which comprise the a) components shown in Table 1 was observed in the same way as in Examples 1 to 14, and the results are shown in Table 1.

As is clear from the results in Examples 1 to 14, by incorporating the b) component there is a considerable improvement in the heat resistance.

Hence, these curing compositions can be said to be useful as the main component of a hot-melt adhesive with excellent heat resistance.

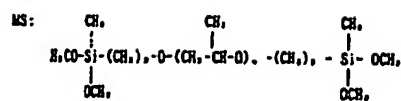
Table 1

	a) Component		b) Component		Silanol Catalyst		Softened State
	Type	Parts	Type	Parts	Type	Parts	
Ex.1	Asahimelt #600	500	MS	100	OT/LA	1/0.25	Δ
Ex.2	Asahimelt #600	200	MS	100	OT/LA	1/0.25	O
Ex.3	Asahimelt RS45	500	PTHF	100	OT/LA	1/0.25	Δ
Ex.4	Asahimelt RS45	100	PTHF	100	OT/LA	1/0.25	O
Ex.5	Meltlon E802	500	MVL	100	OT/LA	1/0.25	Δ
Ex.6	Meltlon E802	200	MVL	100	OT/LA	1/0.25	O
Ex.7	Asahimelt F085	500	PCL	100	OT/LA	1/0.25	O
Ex.8	Asahimelt F085	200	PCL	100	OT/LA	1/0.25	O
Ex.9	Balloonmelt B3000	500	PCL	100	OT/LA	1/0.25	Δ
Ex.10	Balloonmelt B3000	200	PCL	100	OT/LA	1/0.25	O
Ex.11	Vistanex LM-MS	500	PIB-1	100	OT/LA	1/0.25	Δ
Ex.12	Vistanex LM-MS	200	PIB-1	100	OT/LA	1/0.25	O
Ex.13	Hamatite PRC-488-Y	500	PIB-2	100	OT/LA	1/0.25	O
Ex.14	Hamatite PRC-488-Y	200	PIB-2	100	OT/LA	1/0.25	O
Comp.1	Asahimelt #600	100	-	-	-	-	X
Comp.2	Meltlon E802	100	-	-	-	-	X
Comp.3	Asahimelt F085	100	-	-	-	-	X
Comp.4	Balloonmelt B3000	100	-	-	-	-	X
Comp.5	Vistanex LM-MS	100	-	-	-	-	X
Comp.6	Hamatite PRC-488-Y	100	-	-	-	-	X

O = retains shape and no tack, Δ = retains shape but tacky, X = melts
(Notes)

Asahimelt #600 and RS45 (vinyl copolymer resin made by Asahi Chemical Industry Co.)
Meltlon E802 (polyester hot-melt adhesive produced by Diabond Kogyo K.K.)
Asahimelt F085 (polyurethane hot-melt adhesive made by Asahi Chemical Industry Co.)
Balloonmelt B3000 (polyolefin hot-melt adhesive made by Asahi Chemical Industry Co.)
Vistanex LM-MS (polyisobutylene of MWt 8700-10,000 produced by Exxon Chemical)
Hamatite PRC-488-Y (hot-melt butyl filler formula made by Yokohama Rubber)
OT = tin octylate LA = laurylamine

Table 1 (continued)



$M_n \approx 8,000$ $P_n(51) \approx 1.4 - 1.8$

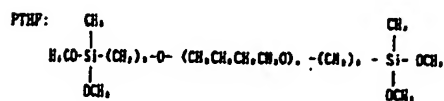
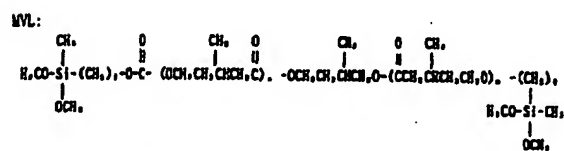
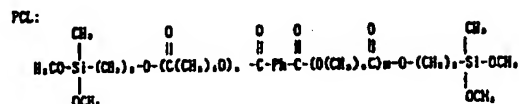
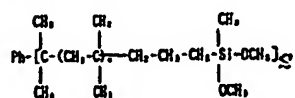
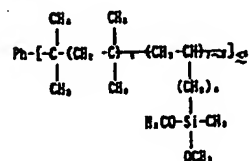

$$M_D \approx 4,000 \quad F_D(SI) \approx 1.4 \sim 1.8$$

$$M_n \approx 4.000 \quad P_n(SI) \approx 1.3 \sim 1.7$$

Table 1 (continued)


$$M_n \approx 12,000 \quad P_n(Si) \approx 1.4 \sim 1.8$$

$$M_n \approx 5,000 \quad P_n(SI) \approx 2.0 \sim 2.5$$

$$M_D \approx 8,000 \quad P_D(SI) \approx 2.5 \sim 3.5$$

Example 15

Curing compositions (A, B, C and D) were prepared by mixing together 100 parts by weight of PIB-1 as the b) component and 0, 20, 50 or 200 parts by weight of Vistanex LM-MS as the a) component, plus 1/0.25 parts of OT/LA as the silanol catalyst and 1 part by weight of water, after which in each case the mixture was cast at 50°C into a tubular moulding frame and then hardening/curing carried out for 1 week at 50°C.

Using the curing compositions obtained in this way (cylindrical sample: diameter 8 mm and height 6 mm) measurement of the viscoelastic properties was carried out (at 10 Hz, and a heating rate of 2°C per minute) in compression mode (0.5% strain).

The measurement of the viscoelastic properties was performed by means of a Rheometrics RS.

The results obtained are shown in Figures 1, 2 and 3.

Sample No.	PIB-1	Vistanex LM-MS	Curve
A	100	0	_____
B	100	20	_____
C	100	50	_____
D	100	200	_____

As is clear from Figures 1 to 3, there is no lowering of the elastic modulus at high temperatures in the system where 200 parts by weight of the Vistanex LM-MS was added, and indeed it was actually increased. Furthermore, in the systems where 50 to 200 parts by

weight of Vistanex LM-MS was added, when compared to the system where no addition was made a high tan δ value was shown over a broad range, demonstrating value as a vibration-deadening/damping material.

Examples 16-18 and Comparative Examples 7 and 8

Curing compositions were prepared by carrying out compounding as shown in Table 2 and then, after three passes using a triple roll paint mill, the mixture in each case was used to fill a moulding frame of thickness 3 mm, following which hardening and curing were carried out for 1 week at 50°C. Thereafter, dumbbells [JIS K7113 No.2(1/3) dumbbells] were punched out and the temperature dependency of the dumbbell properties measured.

The results are shown in Table 3.

Table 2

Composition Formula	Example 16	Example 17	Example 18	Comparative Example 7	Comparative Example 8
PIB-1	100	100	100	100	0
Vistanex LM-MS	100	100	50	0	100
polybutene	75	95	75	55	55
CCR	140	280	140	140	140
TiO ₂	7	14	7	7	7
Tinuvin 327	1	2	1	1	1
Sanol 770	1	2	1	1	1
Irganox 1010	1	2	1	1	1
Na ₂ SO ₄ ·10H ₂ O	5	5	5	5	5
CB #45	11.25	11.25	11.25	11.25	11.25
OT	3	3	3	3	3
LA	0.75	0.75	0.75	0.75	0.75

(Notes)

Polybutene (Idemitsu Polybutene OH)

CCR (Hakuenka [calcium carbonate] produced by Shiraishi Kogyo K.K.)
 TiO₂ (R-820, produced by Ishihara Sangyo K.K.)
 Tinuvin 327, Irganox 1010 (produced by Ciba Geigy)
 Sanol 770 (produced by Mitsui)
 Na₂SO₄.10H₂O (produced by Wako Pure Chemical Industries)
 CB #45 (produced by Mitsubishi Kasei Corp.)

Table 3

		-20°C	0°C	23°C	50°C	100°C	150°C	200°C
Example 16	M ₁₀	1.1	0.3	0.1	0.3	0.2	0.3	0.4
	M ₅₀	3.1	1.3	0.8	0.9	0.9	1.0	1.4
	M ₁₀₀	4.5	2.2	1.5	1.7	1.6	-	2.7
	TB	14.8	7.4	4.0	2.5	1.6	1.4	2.9
	EB	524	417	319	196	100	82	110
Example 17	M ₁₀	1.5	0.8	0.3	0.8	0.4	0.7	0.3
	M ₅₀	4.5	2.5	1.6	2.1	1.5	2.3	2.2
	M ₁₀₀	7.3	4.6	3.4	3.7	3.3	4.4	-
	TB	14.4	9.5	5.2	5.1	4.0	4.7	2.8
	EB	410	339	207	186	134	114	69

Table 3 (Continued)

		-20°C	0°C	23°C	50°C	100°C	150°C	200°C
Example 18	M ₁₀	1.1	0.5	0.3	1.5	0.4	0.6	0.9
	M ₅₀	2.7	1.9	1.2	2.5	1.4	2.1	2.5
	M ₁₀₀	4.4	3.2	2.2	3.8	2.8	3.9	4.3
	TB	21.8	14.2	5.5	6.7	3.9	5.5	4.5
	EB	521	517	308	270	165	159	107
Comparative Example 7	M ₁₀	2.7	1.3	0.9	-	1.2	1.3	1.4
	M ₅₀	6.1	4.4	3.1	-	3.6	4.7	5.3
	M ₁₀₀	9.3	7.4	5.4	-	5.7	7.5	8.5
	TB	37.5	26.2	15.1	-	6.6	10.0	9.4
	EB	468	450	381	-	134	182	126

(Notes) units of M₁₀, M₅₀, M₁₀₀ and TB = kg/cm², and for EB = %

In the case of Comparative Example 8, there was considerable tackiness at room temperature and it was impossible to punch-out dumbbells.

Amongst the results in Table 3, the data for TB and EB are shown graphically in Figure 4, taking the value at 23°C as 1.

From Figure 4, if a comparison is made between the extent of lowering of TB and EB at high temperatures in the case of the curing composition not containing any a)

component hot-melt resin (Comparative Example 7) and in the case of the curing compositions where 50-100 parts by weight of the a) component hot-melt resin was introduced, it is clear, from the fact that there was no great difference, that the heat resistance of the component a) (on the high temperature side) is markedly improved by the introduction of component b).

The composition (Comparative Example 8) which did not contain any b) component did not of course cure and a strength sufficient to punch-out dumbbells was not achieved.

[Effects of the Invention]

In the curing composition of the present invention, the b) component is blended with the a) component hot-melt resin and, by curing, the composition is suitable as a hot-melt adhesive where the heat resistance, which is the major weakness of the a) component, is simply improved without impairing the various properties inherently possessed by the a) component, such as wetting properties, speed of manifestation of adhesion performance at an early stage, and good usability, etc. Furthermore, in the case where a hot-melt butyl is used as the a) component, by the fact of curing the blended b) component even without vulcanization of the a) component, it is possible to manifest heat resistance due to the b) component and also to manifest a high $\tan \delta$ value on the high temperature side at the unvulcanized viscosity of the a) component, making it suitable as a vibration-deadening/damping material.

4. Brief Explanation of the Drawings

Figures 1 to 3 are graphs showing the results of measurements of the viscoelastic properties of the curing compositions. Figure 4 comprises graphs showing the results of measurements of the temperature dependency of the properties of dumbbells of the curing compositions.

Applicant

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Agents

Patent Attorney Y. Hosoda
(plus 1 other)

Figure 1

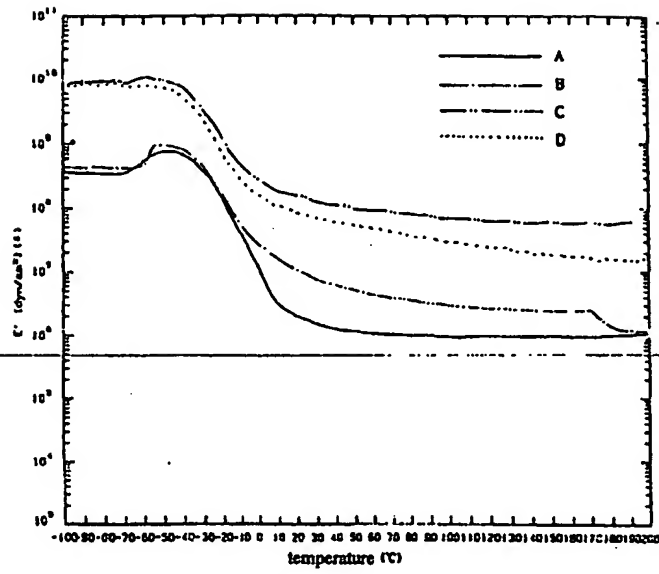


Figure 2

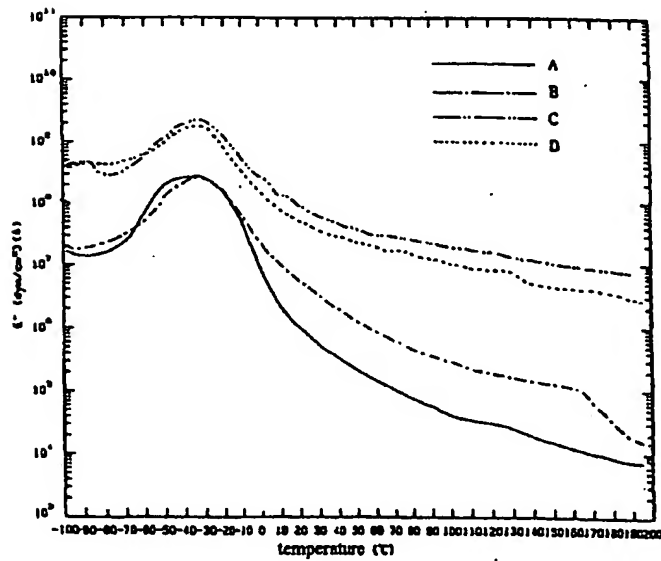


Figure 3

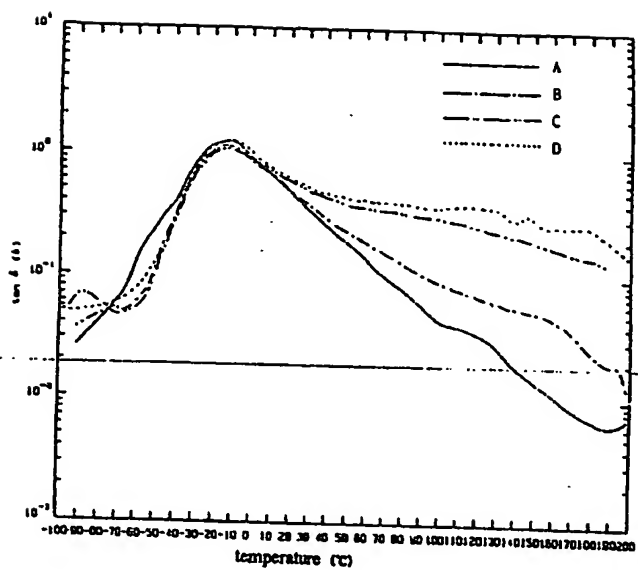
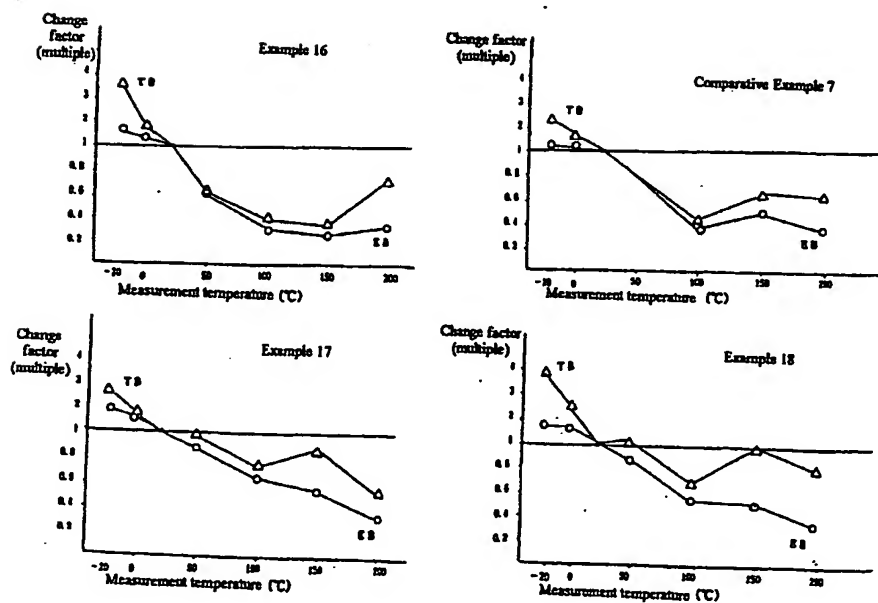


Figure 4



Translator's note

¹ Strictly speaking, the Japanese says "cotton chip" but the meaning of "chip" in such a context is unclear.

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